

## Paper

# INVESTIGATION OF AERIAL DISPERSION OF RADIOACTIVE DUST FROM AN OPEN-PIT URANIUM MINE BY PASSIVE VINYL COLLECTORS\*

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**Abstract**—Detailed investigations of the aerial dispersion of radioactive dust from the biggest open-pit U mining and milling operation in Australia were carried out. Spatial distributions of the long-lived radionuclides of  $^{238}\text{U}$  series and their origin, i.e., mining and milling operations vs. natural background radiation, have been studied. Horizontal flux, dry deposition, and ground resuspension of the radionuclides were investigated along a 50-km transect in the direction of the prevailing monsoonal winds in the region. The study was performed by means of unconventional “sticky vinyl” passive dust collectors, occasionally supported by high-volume air filter samplers. The data from the flux measurements show an inverse square to inverse cubic dependence, and the dry deposition exhibits an inverse square dependence, of radionuclide load vs. distance. The pit has been the predominant contributor of long-lived U series radionuclides to the environment within the radius of several kilometers from the operations. An aerial dispersion computer code (LUCIFER), based on a Gaussian plume model, was developed for the project. Experimental data were used as the code input data. Good agreement between the measured data and the normalized computed results was obtained.

## INTRODUCTION

THE BIGGEST U mining and milling operation in Australia, Ranger Uranium Mines (RUM), operates an open-pit U mine and ore treatment plant in a remote sub-equatorial part of the Northern Territory. About  $6 \times 10^9$  kg of rock are mined annually from the only operating orebody, no. 1, resulting in an annual output of about  $3 \times 10^6$  kg of uranium oxide ( $\text{U}_3\text{O}_8$ ). The deposits are located about 200 km east of Darwin, in the lowlands of the Alligator Rivers Region, and are surrounded by the UNESCO World Heritage listed Kakadu National Park. Characteristic of the region are two distinct seasons, dry (April–September) and wet (October–March), with steady monsoonal winds.

Earlier investigations indicated that the aerial pathway has been the critical one for transfer of radioactive contaminants from the RUM site to the surrounding environment and the local population (Koperski 1986).

Therefore, detailed investigations of the aerial dispersion of radioactive dust were undertaken.

Investigations were conducted between mid-1984 and the end of 1986. At that time about 1500 people lived permanently near RUM, among them about 1200 in Jabiru township, 8 km west-northwest of Ranger, and about 300 in Jabiru East township, 3 km northwest of Ranger (Fig. 1). The Ranger site and the townships are located along the path of the winds, with the prevailing dry season winds blowing toward the townships (see Figs. 1 and 3).

The aims of the study were: (1) to investigate the spatial distribution of the long-lived radionuclides of the  $^{238}\text{U}$  series along the path of the monsoonal winds in the region and (2) to investigate the origin of the radionuclides, i.e., mining operations vs. natural radiation background sources.

The rationale behind the use of the “sticky vinyl” type of passive collectors was, first, their capacity to distinguish between different directions of airborne disper-

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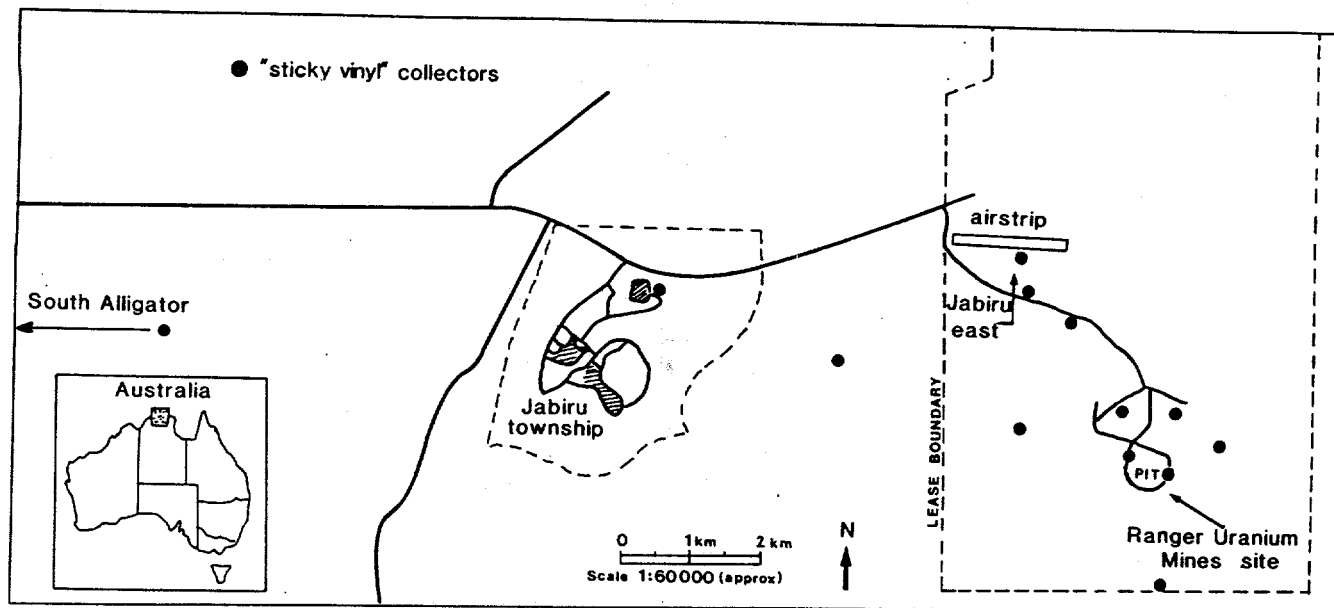


Fig. 1. Sampling locations.

sion, in contrast to traditional active air samplers; second, their collection potential of dry deposited radionuclides; and third, their low material and operational cost and their ease of installation in the field. The latter are important factors when investigations are conducted in places where provision of power necessary to operate active samplers is difficult or practically impossible to obtain.

### MATERIALS AND METHODS

The use of passive deposition collectors to indicate the presence and levels of atmospheric pollutants has been described earlier. Both man-made indicators such as tackyshades, dry cloth, and rain collectors (Eubank and Ward 1964; Fry 1982; Makhonko 1964; McHugh et al. 1986), and bioindicators like naturally growing lichen (Pettersson et al. 1988) have proven to be useful.

The passive "sticky vinyl" dust collectors used in this study employed a domestic-type, clear, self-adhesive vinyl.<sup>‡</sup> A vinyl sheet is covered on one side by a thin layer of sticky substance. The other side of the vinyl sheet is non-sticky, smooth, and easily washable. Each collector consisted of a sheet of vinyl stretched over a plywood frame with areas between 0.35 and 1 m<sup>2</sup>. Horizontal frames were placed at a height of 1 m and the vertical frames 1.5 m above the ground at their center. The vertical vinyl collection surfaces faced the pit. The monitoring sites extended along a 50-km transect from the Ranger site, parallel to the path of the prevailing winds in the region, with additional sites in other directions. The latter were chosen in order to examine the dependence of

source-to-sampling site direction on dust activity dispersion.

The day-to-day (i.e., short-term changes in source activity releases from the pit area) are large due to mine blasting. The blasting occurs on average five times per week. Therefore, in order to determine the average long-term activity dispersion from the mine site, relatively long sampling periods were chosen. Collection of airborne dust by means of vinyl collectors was conducted for a total of 240 d during the 3-y period. The total collection time during the first year (1984) was 100 d. The collectors were replaced about once a month.

During one monthly exposure, additional horizontally oriented vinyl collectors were set up at 1 m height, with the collection sides facing the ground, to assess re-suspension of radioactivity from the ground at the sampling sites. In addition, surface soil was also sampled at the same monitoring sites by stripping the top millimeter (approximately) of the soil cover from a surface area of about 1 m<sup>2</sup>.

Dust load and its retention on the vinyl collectors were studied separately. A single vertical vinyl collector, divided into eight identical strips of 0.35 m<sup>2</sup> each, was gradually exposed in weekly intervals during an 8-wk-long exposure. This experiment took place at a sampling site close to the pit (about 400 m from the pit center).

Simultaneous active dust sampling was conducted periodically at some of the passive collector sites by means of a high-volume air sampling technique. The latter has been used to facilitate determinations of dry deposition velocities.

Analyses of radionuclide concentrations on vinyl collectors, air filters, and in soil were carried out by both  $\alpha$  spectrometry and  $\gamma$  spectrometry. The applied methods of radiochemical sample treatment and radionuclide sep-

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ation have been reported elsewhere (Holm 1984; Martin et al. 1985). The specific activities of the dust from the vinyl collectors were determined by gravimetric measurements on the mass load per unit area of the horizontal vinyls.

## AERIAL DISPERSION MODEL

### Theory

The problems of predicting the dispersion of airborne material released from a source are commonly approached by solving the diffusion-transport equation. The Gaussian plume equation for a point source was formulated by Pasquill (1961):

$$C(x, y, z) = \frac{Q}{2\pi\sigma_y\sigma_z\mu} \exp\left[-\left(\frac{y}{\sigma_y}\right)^2/2\right] \times \left[ \exp\left[-\left(\frac{z-H}{\sigma_z}\right)^2/2\right] + \exp\left[-\left(\frac{z+H}{\sigma_z}\right)^2/2\right] \right], \quad (1)$$

where

$C$  = concentration of the gas or particles at position  $x, y, z$ ;

$x$  = distance from the source in the downwind direction (m);

$y$  = distance from the source in the crosswind direction (m);

$z$  = height above the ground (m);

$Q$  = source activity release rate ( $\text{Bq s}^{-1}$ );

$\sigma_y$  = standard deviation of the horizontal Gaussian distribution (m);

$\sigma_z$  = standard deviation of the vertical Gaussian distribution (m);

$\mu$  = wind speed ( $\text{m s}^{-1}$ ); and

$H$  = the effective release height (m).

The third exponential term considers the reflection of gases at the ground and was subsequently omitted when particulates were treated. The dispersion coefficients  $\sigma_y$  and  $\sigma_z$ , which are dependent on atmospheric stability conditions, are evaluated from Briggs equations (Briggs 1974).

Depletion of the concentration in the plume may be the result of dry or wet deposition. Except for scavenging by rain, dry deposition accounts for all those processes by which airborne matter may be removed from the plume. The removal is caused mainly by gravitational settling, chemical and physical impaction of the diffusing plume on the underlying ground surface, and solution in surface waters. The dry deposition rate is given by (Whicker and Schultz 1982):

$$R_d(x, y) = V_d \times C(x, y, 0), \quad (2)$$

where

$R_d$  = the dry deposition rate ( $\text{Bq m}^{-2} \text{s}^{-1}$ );

$V_d$  = the dry deposition velocity ( $\text{m s}^{-1}$ ); and

$C(x, y, 0)$  = the ground level plume activity concentration ( $\text{Bq m}^{-3}$ ).

The wet deposition rate is given by:

$$R_w(x, y) = \phi \times \int_0^\infty C(x, y, z) \times dz, \quad (3)$$

where

$R_w$  = the wet deposition rate ( $\text{Bq m}^{-2} \text{s}^{-1}$ ) and  
 $\phi$  = wet deposition coefficient ( $\text{s}^{-1}$ ).

### Computer code

A computer code, LUCIFER (Erlandsson and Pettersson 1988), based on the Gaussian plume model, was developed for this project. The code was designed to calculate the air concentration and the dry and wet deposition rates of radionuclides as a function of distance from the area source.

When treating aerial dispersion from large area sources, the air concentration and deposition rate calculations become quite complicated. When designing the computer code, the large area source (the mine pit) was treated as a finite number of point sources, which were analytically integrated in the  $x$ - and  $y$ -directions. The area source was assumed to be homogeneous and circular, the latter being a fair approximation of the shape of the mine pit. Therefore, the area source is regarded as a series of line sources confined in a circle. Although there are different ways of treating an area source problem (Moore et al. 1979; Lewellen et al. 1985), the main difference in the LUCIFER code is that it considers contributions from the source to a point of interest from all wind directions. The distribution, including depletion by dry deposition, is first calculated for 16 sectors of  $22.5^\circ$  each. Then the contribution from each sector to the center line for the sector of interest is added, taking into account the different sector wind frequencies. The final sector average air concentration for the exposure period of interest,  $\bar{C}(x)$ , is calculated on the basis of the time average of the input parameters,  $\bar{\mu}$ ,  $\bar{\sigma}_y$ ,  $\bar{\sigma}_z$ .

### Air concentration vs. vinyl activity load

The activity load on vertical vinyl collectors,  $J(x)$ , does not represent air concentration directly but is correlated to the sector average of the directional horizontal flux,  $\bar{I}(x)$  ( $\text{Bq m}^{-2} \text{d}^{-1}$ ) and air concentration,  $\bar{C}(x)$  ( $\text{Bq m}^{-3}$ ), by:

$$J(x) = \alpha \times \bar{I}(x) = \frac{\alpha \times L \times C(x)}{t}, \quad (4)$$

where

$\alpha$  = constant including the vinyl collector collection and retention efficiencies of the radionuclides;

$L$  = total air column passing the vinyl collector in the direction from the source (m); and

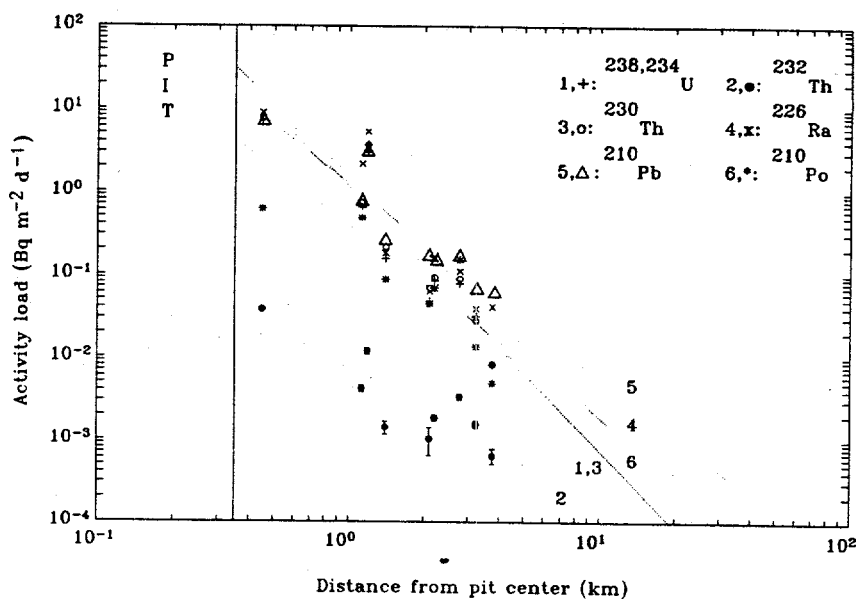
$t$  = exposure time (d).

## RESULTS AND DISCUSSION

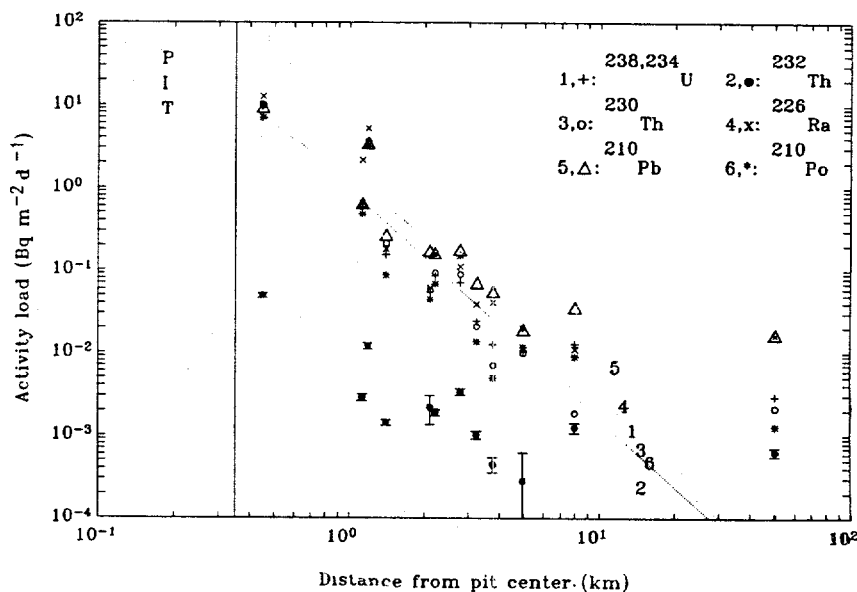
*Activity load on vertical vinyls*

Results of monitoring airborne, long-lived radionuclides by means of the vertical vinyl collectors are shown in Fig. 2a and 2b for both single dry season (1984) and the combined three dry seasons (1984–1986), re-

spectively. The results reflect the dry season radionuclide activity loads on the vinyl collectors in absolute terms. At the same time, the data also reflect the dry season average activity flux distribution of radionuclides by the wind blowing along the directions linking the pit area with the sampling sites (see eqn 4). To obtain these values, the average wind frequency in a 22.5° wind sector around



(a)



(b)

Fig. 2. Dust activity load (arithmetic means) on vertically oriented "sticky vinyl" collectors during one (1984) dry season (Fig. 2a) and during three (1984–86) dry seasons (Fig. 2b) ( $\pm 1$  SD for  $^{232}\text{Th}$  is shown; for the other radionuclides,  $\pm 1$  SD  $< 5\%$ ). The dotted lines represent the best fit to the data, eqn (5).

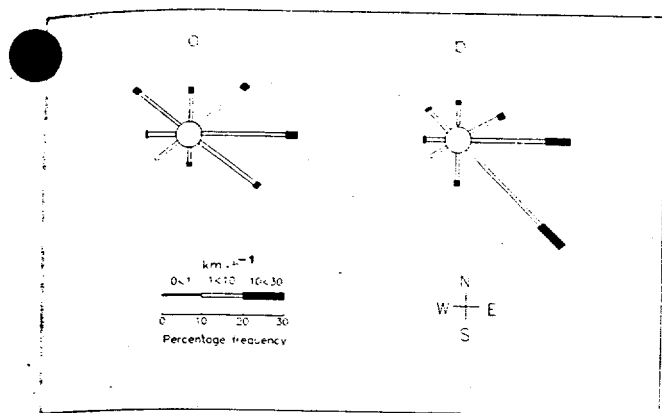


Fig. 3. Wind roses for the study periods: a) single dry season (1984), b) combined three dry seasons (1984-1986).

each sample site was first determined by examining RUM's meteorological data from the study area. Second, the activity load data were normalized to 100% wind frequency in the direction from the source to the sampling site of interest. The latter was done to facilitate a comparison of radionuclide loads in various directions from the source. The wind roses for the single and the combined three dry seasons are shown in Fig. 3.

Based on observations, the decrease in activity load,  $J$ , with the distance from the pit area,  $x$ , may be represented by an equation of the following form:

$$J(x) = k \times x^a, \quad (5)$$

where  $J$  is expressed in  $\text{Bq m}^{-2} \text{d}^{-1}$  and  $x$  in km, and  $k$  and  $a$  are constants for each radionuclide. The lines of best fit using this expression are shown in Fig. 2. The corresponding values for  $k$  and  $a$  are given in Table 1.

No significant difference between  $^{238}\text{U}$  and  $^{234}\text{U}$  data was observed in the entire study; they are therefore given as pooled values. The sampling site at  $x = 50 \text{ km}$  was selected in order to obtain an estimate of the natural background levels in the study area and was omitted in this fitting exercise.

The 3-y average data for the U series radionuclides (Fig. 2b), except  $^{210}\text{Pb}$ , are reasonably well described by an inverse cubic dependence of activity load vs. distance. The lower slope constant for  $^{210}\text{Pb}$  is a consequence of the contribution of atmospheric  $^{210}\text{Pb}$ , originating from airborne  $^{222}\text{Rn}$ . The statistical uncertainty in the individual data is not a significant problem (usually, for 1 standard deviation in counting statistics, less than 5%), but fluctuations in the dust activity generation at the source may create fluctuations in the activity loads at sites far from the source ( $>2-3 \text{ km}$ ). This could be the reason for the deviation in the slope constant,  $a$ , when results from a single-year exposure (Fig. 2a) are compared with the 3-y average exposure (Fig. 2b). The activity load of  $^{210}\text{Po}$  is generally lower than expected, possibly because of evaporation of  $^{210}\text{Po}$  from the vinyl collectors, due to the volatile nature of Po. One would expect a somewhat higher load of  $^{210}\text{Po}$  due to the atmospheric contribution created from airborne  $^{222}\text{Rn}$ , but less than in the case of  $^{210}\text{Pb}$ . When created from airborne  $^{222}\text{Rn}$ , the  $^{210}\text{Po}$ : $^{210}\text{Pb}$  activity ratios in surface air are typically in the range 0.1-0.3 (Jacobi 1979). The activity load of Th-series radionuclides, represented by  $^{232}\text{Th}$ , is much lower than for the U series radionuclides. This was expected because we found that  $^{232}\text{Th}$  activity concentrations in soil are of the same order of magnitude in the pit as in the surroundings (Table 2). The lower slope constant for  $^{232}\text{Th}$  is a result of the proximity to the natural background activity load, and consequently the fit to the curve is very uncertain.

Results from the sampling site located 1.2 km from the pit in the north-northeast direction deviate signifi-

Table 1. The constants  $a$  and  $k$  (eqn 5) for each radionuclide investigated.<sup>a</sup>

Vinyl orientation	Radionuclide	$a_1$	$a_2$	$k_1$	$k_2$	$r_1$	$r_2$
Vertical	$^{238,234}\text{U}$	-3.1	-2.7	1.0	0.9	-0.93	-0.93
Vertical	$^{232}\text{Th}$	-1.7	-1.5	0.007	0.007	-0.85	-0.82
Vertical	$^{230}\text{Th}$	-3.1	-2.7	1.1	1.0	-0.94	-0.91
Vertical	$^{226}\text{Ra}$	-2.9	-2.7	1.5	1.5	-0.91	-0.93
Vertical	$^{210}\text{Pb}$	-2.3	-2.2	1.2	1.2	-0.93	-0.93
Vertical	$^{210}\text{Po}$	-2.3	-2.7	0.4	0.7	-0.76	-0.89
Vertical	Average except $^{232}\text{Th}$ , $^{210}\text{Pb}$	-3.1	-2.7	1.0	1.0	-0.91	-0.91
Horizontal	$^{238,234}\text{U}$		-2.0		11		-0.97
Horizontal	$^{232}\text{Th}$		-1.4		0.07		-0.93
Horizontal	$^{230}\text{Th}$		-2.1		7.9		-0.95
Horizontal	$^{226}\text{Ra}$		-2.2		7.7		-0.93
Horizontal	$^{210}\text{Pb}$		-1.8		13		-0.96
Horizontal	$^{210}\text{Po}$		-2.2		5.2		-0.93
Horizontal	Average, except $^{232}\text{Th}$ , $^{210}\text{Pb}$		-2.1		7.9		-0.95

<sup>a</sup>  $a$  and  $k$  are constants described in the text, and  $r$  is the correlation coefficient. Index 1 corresponds to results in Fig. 2a. Index 2 corresponds to results in Fig. 2b.

Table 2. Specific activities ( $\text{Bq kg}^{-1} \pm 1 \text{ SD}$ ) of radionuclides in dust from dry deposition and of surface soil vs. distance from pit center.

Distance from pit center (km)	Horizontal vinyl			Surface soil		
	$^{238,234}\text{U}$	$^{232}\text{Th}$	$^{230}\text{Th}$	$^{238,234}\text{U}$	$^{232}\text{Th}$	$^{230}\text{Th}$
0	—	—	—	$23,000 \pm 1800$	$115 \pm 7$	$23,000 \pm 700$
0.45	$18150 \pm 600$	$105 \pm 15$	$19900 \pm 1015$	$2150 \pm 85$	$26 \pm 1$	$3285 \pm 140$
1.125	—	—	—	$425 \pm 12$	$38 \pm 3$	$330 \pm 22$
2.775	$1140 \pm 50$	—	—	$115 \pm 4$	$73 \pm 4$	$150 \pm 8$
3.75	$1105 \pm 90$	—	—	$63 \pm 2$	$63 \pm 2$	$79 \pm 2$
4.95	$2090 \pm 50$	$20 \pm 2$	$1890 \pm 40$	$18 \pm 1$	$15 \pm 1$	$20 \pm 1$
7.95	$240 \pm 8$	$13 \pm 2$	$305 \pm 2$	$24 \pm 1$	$42 \pm 1$	$27 \pm 1$
50	$40 \pm 2$	$12 \pm 2$	$38 \pm 3$	$19 \pm 1$	$31 \pm 1$	$17 \pm 1$

cantly from the line of best fit. This site is close to the ore treatment plant, which may result in activity flux levels from the plant being significant compared to the activity flux levels from the pit at this sampling site. The plant contribution to the total aerial activity flux from the mining area has not been investigated in this project.

When comparing 1- and 3-y average exposure data, one can see that both the magnitude of the activity flux from the source and the dispersion pattern do not change substantially during the dry season or over several dry seasons. Only the occasional short-term monsoonal rainstorms will alter the source term.

One particular problem associated with the use of the "sticky vinyl" collectors is the collection of not only dust but also insects, mainly flies and mosquitos, and forest litter. This may ultimately result in an increased amount of radionuclides on the vinyl collectors. One site, situated 2.8 km in a northwest direction and close to a road lamp, collected considerable quantities of insects and showed generally raised  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  activity values.

#### Dry deposition on horizontal vinyls

Average dry deposition activity concentrations, for the combined three dry seasons (1984–1986), of the U series radionuclides and  $^{232}\text{Th}$  as a function of distance from the pit are shown in Fig. 4. The data are normalized to 100% wind frequency in the direction from the source to each sampling site. A fit to the data, using the same expression as for the vertical collectors, gives a somewhat slower rate of decrease with distance (Table 1). This was to be expected due to contributions to the dry deposition from sources in directions other than that of the pit area, i.e., natural background sources. Here again,  $^{210}\text{Pb}$  exhibits a slower slope constant than the other U series radionuclides. This is partly due to the different plume dispersion profile (Fig. 2a, b), but the main reason is the different activity size distribution, which is discussed later in the text.

Formally, it is not correct to normalize the data to wind frequencies in this case, especially if contributions to dry deposition from sources other than the pit are sig-

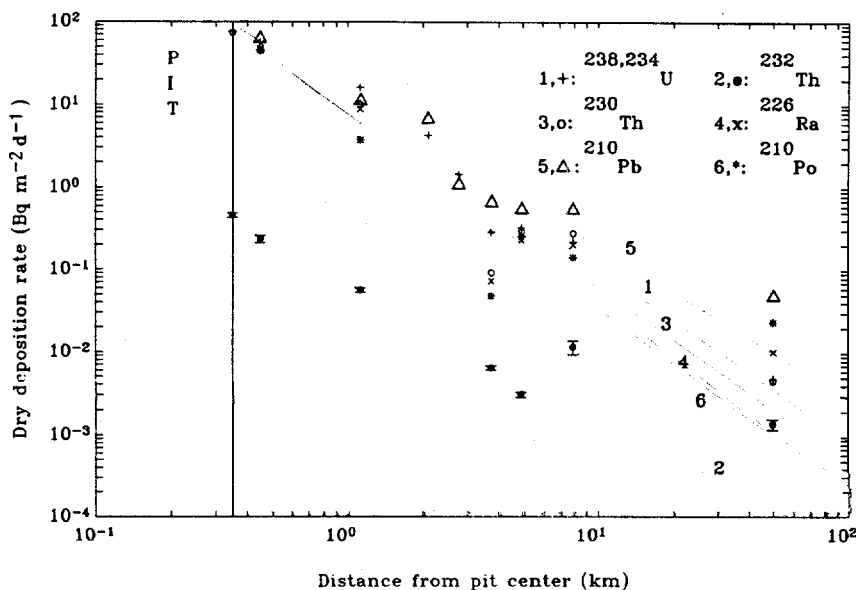


Fig. 4. Dry deposition rate (arithmetic means) on horizontally oriented "sticky vinyl" collectors during three dry seasons ( $\pm 1 \text{ SD}$  for  $^{232}\text{Th}$  is shown; for the other radionuclides,  $\pm 1 \text{ SD} < 5\%$ ).

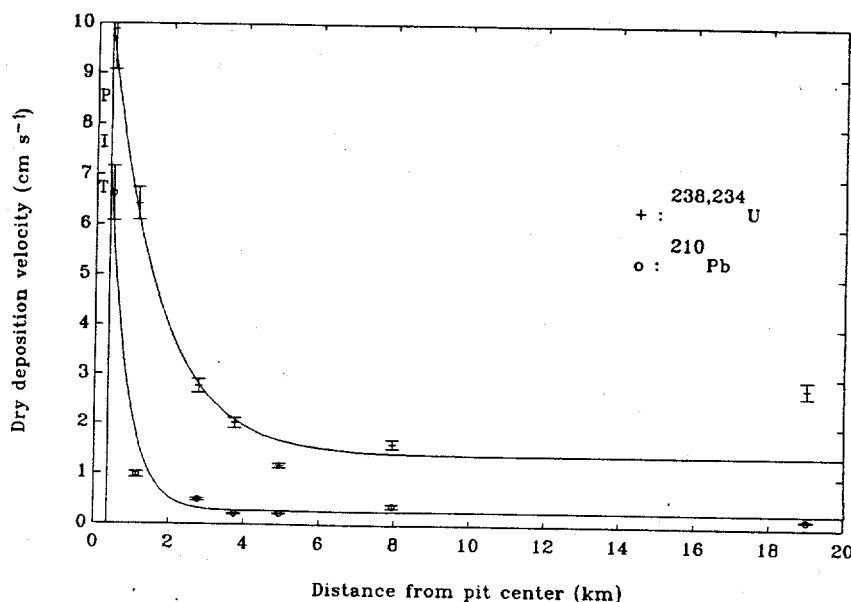


Fig. 5. Dry deposition velocity distributions ( $\pm 1$  SD) with "sticky vinyl" as collection substrate.

nificant, which might be the case for the sampling points far from the pit. Such an approach, however, was necessary to enable a comparison of the dry deposition data from sampling sites in various directions.

During two exposure periods of 4 wk each, high-volume air samplers were also used at seven of the vinyl collector locations. The average activity concentration in the air 1 m above the ground was thereby obtained and the dry deposition velocities,  $V_d$ , were determined (eqn 2). Figure 5 shows  $V_d$  as a function of distance from the pit center for  $^{238,234}\text{U}$  and  $^{210}\text{Pb}$ . The solid lines represent the best exponential fit to the data (the 50 km sampling site data excepted), given by the expressions:

$$V_d = 0.014 + 0.086 \exp[-0.75(x - r)], \quad \text{for } ^{238,234}\text{U}$$

and

$$V_d = 0.0027 + 0.07 \exp[-2.0(x - r)], \quad \text{for } ^{210}\text{Pb} \quad (6)$$

where  $x$  = distance from the pit center (km) and  $r$  = source (pit) radius (km) = 0.35 km.

The continual decrease in  $V_d$  from the mine area up to a distance of about 5 km reflects a gradual change in the particle activity size distribution of the dust dispersed from the mining area. The larger particles will deposit in preference to the smaller ones, thus gradually changing the activity size distribution. The difference in  $V_d$  between  $^{238,234}\text{U}$  and  $^{210}\text{Pb}$  indicates the following. First, the source activity size distributions of  $^{210}\text{Pb}$  and U are different, which is likely due to the creation of  $^{210}\text{Pb}$  by decay of  $^{226}\text{Ra}$  and thereby a different physicochemical form of  $^{210}\text{Pb}$  in the solid matrix. Second, away from the source, the  $^{210}\text{Pb}$  produced in the atmosphere will contribute significantly to the air concentration of  $^{210}\text{Pb}$  but to a lesser extent to the dry deposition, due to the relatively small

particles (deduced from the deposition velocities) (Styro and Shalveyus 1966), resulting in a much faster decrease in  $V_d$  than that for  $^{238,234}\text{U}$ .

The range of  $V_d$ s obtained for the vinyl collectors is in agreement with literature data for similar "sticky" substances (Convair 1960; Makhonko 1970) but higher than that which is common for vegetation and bare soil (Sehmel 1980). It is likely that the sticky surface more easily retains particulates than do vegetation and soil.

The specific activities on the horizontal vinyls together with surface soil activity concentrations are shown in Table 2. It is interesting to note the close agreement between surface soil activity concentration in the pit, which corresponds to about 0.18% U, and the dry deposition concentration close to the pit, despite the fact that the pit sample represented only a small surface grab sample. In the surroundings more than a few kilometers away from the pit, the dry deposition specific activities of U series radionuclides clearly exceed the specific activities for surface soil. The observed phenomenon clearly indicates a significant activity contribution of U series radionuclides from the pit to the top soil of the area within the distance of several kilometers from the pit.

#### Resuspension of radionuclides from the ground

To assess the degree of resuspension of dust activity from the ground at a sampling site, a "vinyl resuspension coefficient,"  $R_v$ , has been defined as the ratio of radionuclide concentration on a vinyl facing downwards to that facing upwards. This gives a comparison between ground resuspension and dry deposition in qualitative terms. Ranges of  $R_v$  values obtained are shown in Table 3. As it is likely that vinyl collectors exposed in these two directions had similar dust particle attachment and retention properties, one may conclude that ground resus-

Table 3. Vinyl resuspension coefficient,  $R_v$ .

Radionuclide	$R_v$ (range)
$^{238,234}\text{U}$ , $^{230}\text{Th}$	0.01–0.035
$^{210}\text{Pb}$	0.004–0.03
$^{210}\text{Po}$	0.02–0.03
$^{232}\text{Th}$	0.03–0.06

pension of radionuclides is of minor importance compared with the contribution from the mine site.

#### Load saturation on vinyl collectors

Load and retention capacities of the vinyl collectors are shown in Fig. 6 as cumulative activity and dust load vs. exposure time. The amount of air (m of air column) passing each stripe during respective exposure intervals was derived from meteorological data by including all wind directions and their frequencies in the path from the source to the vinyl strips. The cumulative activity loads of  $^{238,234}\text{U}$  and  $^{210}\text{Po}$  are close to linear ( $r = 0.9988$  and  $r = 0.9986$ , respectively). The load of  $^{210}\text{Po}$  is lower, as explained earlier. The non-zero intercept implies that the vinyl acted differently at the beginning of the exposure. This is clearly seen from the cumulative dust load curve. No evident explanation of this effect was found. One may suspect that in the tropical heat of the region, some volatile components of the sticky substance evaporate during the initial period of heat exposure. This hypothesis has not been tested under field conditions but indications of such behavior have been observed in laboratory tests. Because of this, uncertainties arose when determining tare weights per unit area for calculations of specific activity (Table 2). To minimize this problem, we used the total weight per unit area of the vinyl with least specific activity found during all three seasons as tare weight.

#### $^{230}\text{Th}$ : $^{232}\text{Th}$ activity ratios

One of the possible ways to determine the impact of aerial dispersion of long-lived radionuclides from an open pit U mine to the environment is to compare activity ratios of selected radioisotopes from the U and the Th series at the points of interest relative to the ratios typical for the pit. In this context, the activity ratios of  $^{230}\text{Th}$  to  $^{232}\text{Th}$  have been examined in the dust samples collected by means of the vertical and horizontal vinyls, as well as in soil samples. Results of the relevant calculations are given in Fig. 7. The activity ratios for the vinyl samples are calculated as arithmetic means of the individual activity ratios during the 3-y sampling, and surface soil data were taken from Table 2.

The  $^{230}\text{Th}$ : $^{232}\text{Th}$  activity ratios in the dust samples from both vertical and horizontal vinyl collectors exhibit much slower decrease in magnitude with distance than the corresponding ratios found in the soil samples. Even at points several kilometers from the pit, there is at least one order of magnitude difference between the vinyl and the soil activity ratios. This further confirms the predominance of the pit area as a source of the airborne, long-lived radionuclides in the region adjacent to the operations.

#### COMPUTER CODE RESULTS VS. EXPERIMENTAL RESULTS

The necessary input parameters for the LUCIFER program were chosen as follows:

- 1) Average wind speed ( $\bar{\mu} = 2.35 \text{ m s}^{-1}$ ) and frequency in 16 sectors of  $22.5^\circ$  each; extracted from meteorological data.
- 2) Dry deposition velocities, according to results in Fig. 5.

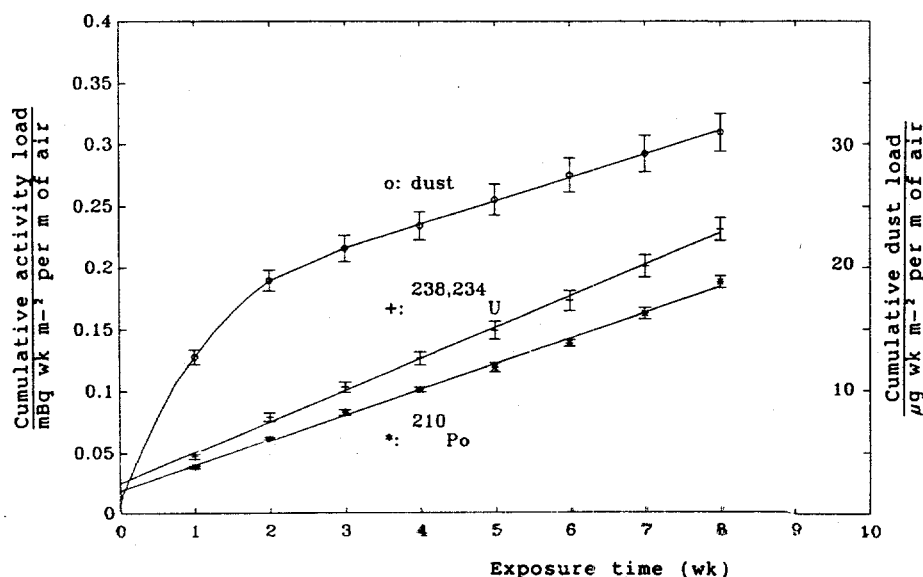


Fig. 6. Cumulative activity and dust load ( $\pm 1$  SD) with "sticky vinyl" as collection substrate.



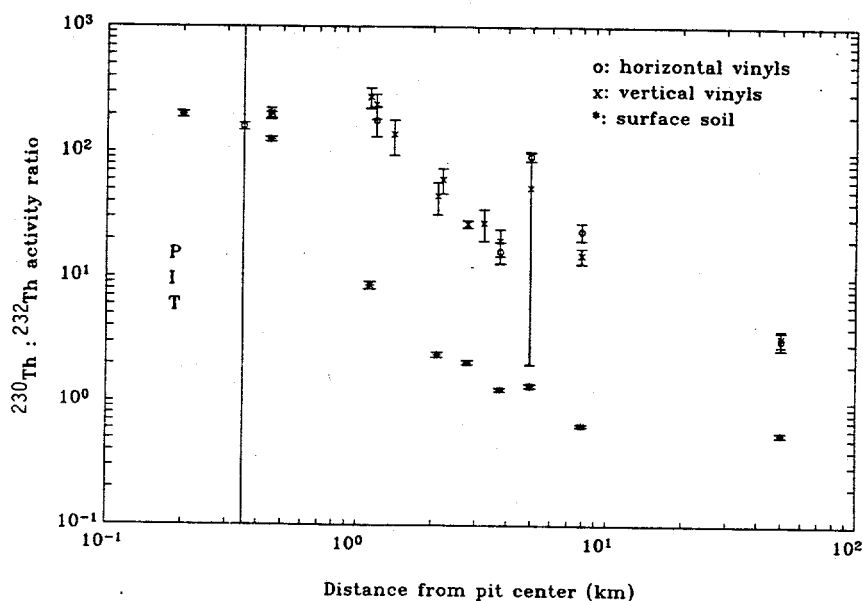


Fig. 7.  $^{230}\text{Th} : ^{232}\text{Th}$  activity ratios (arithmetic means  $\pm 1$  SD) for the three dry seasons: horizontal "sticky vinyl" collectors (dry deposition), vertical "sticky vinyl" collectors, and surface soil.

3) Classification of the region's weather type according to Pasquill categories (Pasquill 1961), based on a) meteorological data for 1984–1986 and b) comparison with meteorological data for June–July period of 1989 (Kvasnicka 1990); each of the categories B, C, D, E represented 25% of the total time period.

4) Source activity emission rate; not available at this stage.

Due to the present lack of source activity emission rate, and the knowledge of the magnitude of the constant  $\alpha$  in eqn (4), code results were normalized to experimental data. The most frequent source to sampling direction (northwest) was chosen for the calculation. Figure 8 presents the computer code results compared with experimental results for  $^{238,234}\text{U}$  for the combined three dry seasons (see Fig. 2b). The slope for the experimental data

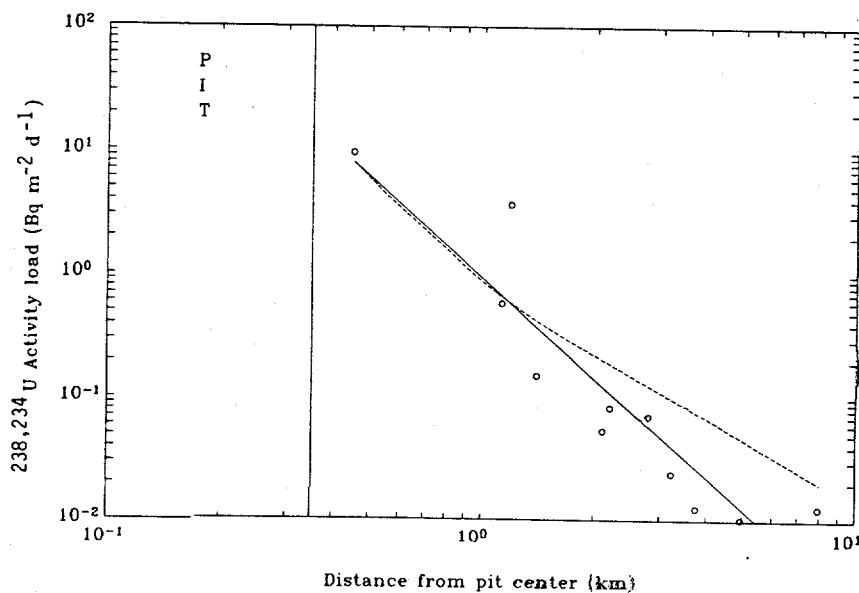


Fig. 8. Comparison between experimental data and computer code (LUCIFER) calculations for the combined three dry seasons. Dashed line: Computer code  $^{238,234}\text{U}$  activity load calculations, normalized to best fit to experimental data (discrete points). Full line: Best fit to experimental data on average  $^{238,234}\text{U}$  activity load on vertically oriented "sticky vinyl" collectors (see Fig. 2b).

was calculated from the best fit according to Table 1. The computer code activity load (air concentration) distribution does not follow the simple power relationship in eqn (5), which is mainly due to the decreasing dry deposition velocity,  $V_d$ , with distance from the source (Fig. 5). The measured  $V_d$  distribution is the sum of the source- and the environment-related  $V_d$ s, which might exaggerate the drop in the source-related dry deposition velocity with distance from the source. The computer code calculations suffer from many uncertainties due to simplifications of the atmospheric conditions, i.e., the use of standard Pasquill stability classes, averaging wind speed, assumption

of zero release height,  $H$ , etc. The latter could markedly deviate from zero due to air turbulence in the pit and blasting. Despite these uncertainties, the computer-code-normalized-activity load agrees fairly well with the experimental results.

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## REFERENCES

- Briggs, G. A. Diffusion estimation for small emissions. Oak Ridge: NOAA, Atmospheric Turbulence and Diffusion Laboratory; Report ATDL-106; 1974.
- Convair. Fission products field release test II, Report NARF-60-10T, U.S. Air Force Nuclear Aircraft Research Facility, Division of General Dynamics Corp., Ft. Worth, TX; 1960. Available from: National Technical Information Service, Springfield, VA.
- Erlandsson, K.; Pettersson, H. B. L. A model for atmospheric dispersion from point and area sources. Presented at Fifth Nordic Radioecology Seminar, Rattvik, Sweden, 1988; Lund, Sweden: Department of Radiation Physics, Lasarettet; 1988 (in Swedish).
- Eubank, B.; Ward, A. Project Shoal. On site health and safety report VUF-1012. Las Vegas, NV: Health and Safety Division, Reynolds Electrical & Engineering Co; 1964.
- Fry, F. A. A comparison of the collection properties of passive samplers and high-volume air samplers for airborne radionuclides. *Radiat. Prot. Dos.* 3:113-115; 1982.
- Holm, E. Review of alpha-particle spectrometric measurements of actinides. *Int. J. Appl. Radiat. Isot.* 35:285-290; 1984.
- Jacobi, W. Lead-210, bismuth-210, polonium-210: Natural activity; internal dosimetry and dose factors for ingestion and inhalation. München, Neuherberg: Institute for Radiation Protection; GSF-Bericht-S-586; 1979 (in German).
- Koperski, J. Radiation exposure of the public from operations of Ranger Uranium Mines. In: *Proceedings of the 4th European Congress and 13th Regional Congress of IRPA*, 15-19 September 1986, Salzburg, Austria; Seibersdorf, Austria: Austrian Society for Radiation Protection; 1986:187-192.
- Kvasnicka, J. Radiation protection check monitoring program for 1989. Darwin, Australia: Mines Environment Directorate, Department of Mines and Energy; 1990.
- Lewellen, W. S.; Sykes, R. I.; Parker, S. F. Comparison of the 1981 INEL dispersion data with results from a number of different models. Springfield, VA: National Technical Information Service; NUREG/CR-4159, ARAP No. 505; 1985.
- Makhonko, K. P. Diurnal changes of the dust content of the surface layer of the atmosphere. U.S. Army Foreign Science and Technology Center, FSTC 381-T64-47; 1964. Translation from Russian (source unknown). Available from: U.S. Army Foreign Science and Technology Center, Washington, DC 20315.
- Makhonko, K. P. The deposition of radioactive dust and its elimination from the atmosphere by precipitation. Springfield, VA: National Technical Information Service; USSR Reports on Natural and Fallout Radioactivity, AEC-tr-7128; 1970.
- Martin, P.; Pettersson, H. B. L.; Hancock, G. Measurement of uranium and thorium series radionuclides by alpha spectrometry. In: *Measurements of long-lived environmental radionuclides, proceedings of a workshop*, 29 April to 1 May 1985, Sydney, Australia. Available from: Office of the Supervising Scientist, Jabiru, N.T. 0886, Australia.
- McHugh, J. O.; Smith, B. D.; Hunt, G. J.; Thomas, R. E. G. The MAFF dry cloth collector programme for monitoring airborne radioactivity. *J. Soc. Radiol. Prot.* 6:63-67; 1986.
- Moore, R. E.; Baes, C. F., III; McDowell-Boyer, L. M.; Watson, A. P.; Hoffman, F. O.; Pleasant, J. C.; Miller, C. W. AIRDOS-EPA: A computerized methodology for estimating environmental concentrations and dose to man from airborne releases of radionuclides. Oak Ridge, TN: Oak Ridge National Laboratory; ORNL-5532; 1979.
- Pasquill, F. The estimation of the dispersion of windborne material. *Meteorol. Mag.* 90:33-49; 1961.
- Pettersson, H. B. L.; Hallstadius, L.; Hedvall, R.; Holm, E. Radioecology in the vicinity of prospected uranium mining sites in a subarctic environment. *J. Environ. Radioactivity* 6:25-40; 1988.
- Sehmel, G. A. Particle and gas dry deposition: A review. *Atmos. Environ.* 14:983-1011; 1980.
- Styro, B. I.; Shalaveyus, S. A. Deposition rate of RaD in the atmosphere. In: *Proceedings of the Atmospheric Scavenging of Radioisotopes*, 7-9 June 1966, Palanga, Russia. Springfield, VA: National Technical Information Service; 1966.
- Whicker, F. W.; Schultz, V. Radioecology: Nuclear energy and the environment, Vol. 2. Boca Raton, FL: CRC Press; 1982.